

# Resolution of the mystery of counter-intuitive photon correlations in far off-resonance emission from a quantum dot-cavity system

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In a coupled quantum-dot nano-cavity system, the photoluminescence from an off-resonance cavity mode exhibits strong quantum correlations with the quantum dot transitions, even though its autocorrelation function is classical. Using new pump-power dependent photon-correlation measurements, we demonstrate that this seemingly contradictory observation that has so far defied an explanation stems from cascaded cavity photon emission in transitions between excited multi-exciton states. The mesoscopic nature of quantum dot confinement ensures the presence of a quasi-continuum of excitonic transitions part of which overlaps with the cavity resonance.

A quantum dot (QD) coupled to a photonic crystal cavity provides a promising system for studying cavity quantum-electrodynamics (QED) in the solid state [1, 2]. In contrast to their atom-based counterparts, these systems exhibit features that arise from their complex environment. A common effect that surfaced in previous experiments is strong off-resonant emission of a cavity mode (CM) containing one or multiple QDs. Photon correlation measurements revealed that the cavity-mode emission is anti-correlated with the QD excitons at the level of single quanta, proving that *cavity feeding* is mediated solely by a single QD [3, 4]. Surprisingly, however, the photon stream emitted by the far off-resonant CM did not show any significant quantum correlations. Previous experimental [5, 6, 7] and theoretical [8, 9] investigations have focused on explaining cavity feeding in terms of dephasing of the QD excitons mediated either by coupling to acoustic phonons or to free carriers. However, all of the attempts to describe cavity feeding using Markovian dephasing of the fundamental exciton line fail to explain the above mentioned photon correlation signatures that appear to be true for all studied QD cavity-QED systems.

In this Letter, we unequivocally demonstrate that the far off-resonant excitation of the CM is solely due to the mesoscopic nature of quantum dot confinement, which in turn leads to an energetically broad cascaded emission of the QD. In this setting, cavity feeding and its photon correlation signatures can be regarded as an intrinsic feature of QD-cavity systems that arises from the complicated QD multi-exciton level structure. We carry out pump-power dependent photoluminescence (PL) as well as photon auto- and cross-correlation measurements on a nano-structure incorporating a single QD embedded in a photonic crystal (PC) defect cavity [3]. To explain our experimental observations, we develop a new theoretical model for the QD-cavity system, perform numer-

ical calculations of its semi-classical dynamics and compare its predictions with the new experimental findings. While a quantitative comparison between numerical and experimental results is intrinsically difficult, the qualitative agreement we achieve is excellent. In particular, the unusual correlation features found experimentally are naturally reproduced by the model and the simulations.

Before proceeding, we remark that acoustic phonons should contribute to cavity feeding [?] when the CM-exciton detuning does not exceed a cutoff energy determined by the inverse QD size, typically in the range of 1 meV (0.7 nm). In contrast, cavity feeding has experimentally been observed for detunings ranging from +10 meV (−7 nm) to −45 meV (+32 nm) (i.e.  $\sim 70$ –300 CM linewidths). Our focus in this Letter lies on this far off-resonance cavity emission phenomenon, which cannot be explained using (Markovian) pure-dephasing or phonon-assisted processes.

A key aspect of cavity-feeding can be identified from typical PL spectra of a single QD coupled to a PC cavity. Figure 1(a) displays a PL spectrum of such a device on a semi-logarithmic scale with a cavity-mode frequency tuned about 15.3 meV (10.9 nm) red of the  $X^0$  transition using nitrogen deposition [11, 12]. The spectrum was obtained in a liquid-helium flow cryostat by pumping the device at around 838 nm, just below the GaAs bandgap. Even though the QD is driven well below saturation, the CM emission is clearly visible together with the dominant QD PL lines that originate mainly from the neutral exciton  $X^0$ , the negatively ( $X^{1-}$ ) and positively ( $X^{1+}$ ) charged trions, and the biexciton ( $XX^0$ ) transition [13]. We remark here that for our devices and excitation conditions, the magnitude of  $X^0$ ,  $X^{1-}$ , and  $X^{1+}$  are comparable, suggesting that the QD is equally likely to be neutral or charged by a single excess electron or a hole. In addition to these discrete lines, we observe

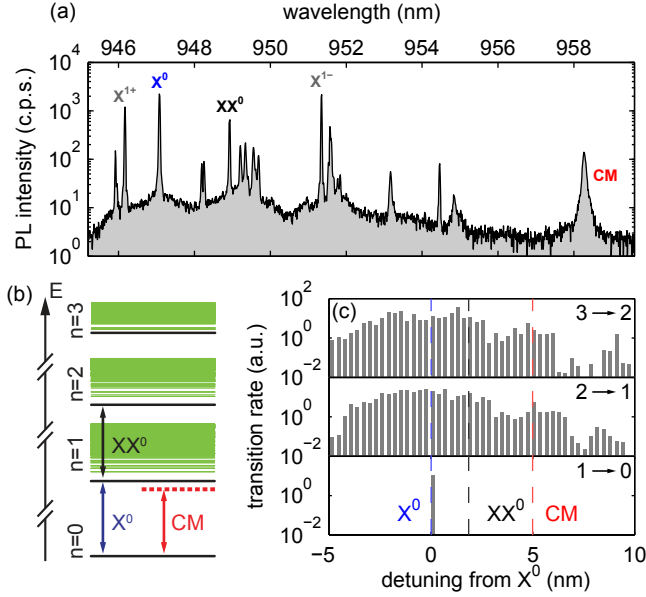


FIG. 1: (color online) QD background emission and level structure. (a) Typical QD PL spectrum on a semi-logarithmic scale. Besides the well-known discrete QD lines, the weak background responsible for cavity feeding is visible. (b) Calculated energy-level diagram ( $n \leq 3$ ) of a neutral QD assuming a truncated parabolic in-plane confinement potential (energy separations between manifolds not to scale). (c) Calculated transition rates between states of different manifolds. States from  $n \geq 2$  manifolds can decay into a large number of final states, thus forming a quasi-continuum of allowed transitions that can feed the CM.

a much broader single-QD background about two orders of magnitude weaker in intensity. The part of this background that is resonant with the CM is Purcell enhanced and leads to the off-resonant cavity luminescence. We emphasize that the principal aim of this Letter is to explain the origin of this single-QD background and how it leads to the observed striking photon correlation signatures.

To understand this background emission, we consider the full excitation spectrum of electron and hole motional states in the QD. While the narrow QD lines originate from states in which the carriers occupy lowest energy single-particle states, there is a large variety of higher excited states for which e.g. a carrier is excited to a p- or d-shell state. This leads to an excitation spectrum consisting of a series of QD manifolds separated approximately by the band-gap energy. We calculate the multi-excitonic eigenstates up to four electron-hole pairs [14, 15] by diagonalizing the Coulomb Hamiltonian, starting from a basis of 18 electron and 32 hole states including spin. The result of the calculation is displayed in Fig. 1(b). In addition, it has recently been demonstrated that higher energy orbital states in an excitation manifold  $n$  (with  $n \geq 2$  electron-hole pairs) are subject to strong hybridiza-

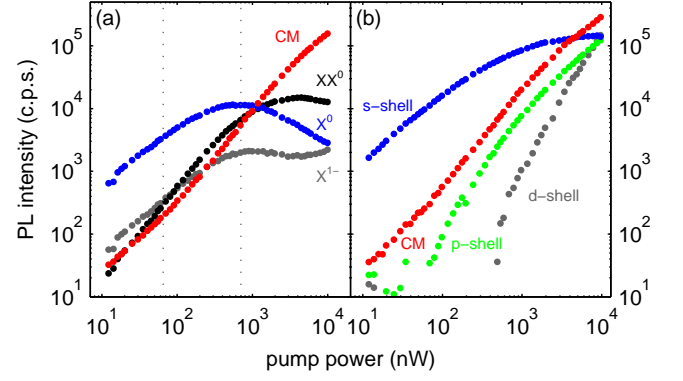


FIG. 2: (color online) Off-resonant cavity emission as a function of pump power. (a) Pump-power dependence of CM,  $X^0$ ,  $X^{1-}$ , and  $XX^0$  emission. The data were obtained by integrating PL intensities for different excitation powers in  $\approx 0.3$  nm wide windows around the different QD lines indicated in Fig 1(a). The CM exhibits complex dynamics due to cavity feeding from higher-lying multi-excitonic states. (b) Comparison of CM emission with the emission from the different QD shells. The CM dynamics is clearly dominated by the QD s-shell for powers below 500 nW, while at higher powers the p-shell takes over.

tion with the wetting-layer continua [16]. This leads to the formation of a continuum of QD excited states for each  $n$ . In this more accurate picture for the QD spectrum, the  $X^0$  line corresponds to a transition from the lowest energy  $n = 1$  manifold state to the ground state ( $1 \rightarrow 0$ ), whereas  $XX^0$  decay relates to a transition between the lowest energy states of the  $n = 2$  and  $n = 1$  manifolds ( $2 \rightarrow 1$ ) [Fig. 1(b)]. Given the large variety of initial and final states,  $2 \rightarrow 1$  (and higher manifold) transitions merge to a quasi-continuum and give rise to QD background emission that feeds the CM [23]. Figure 1(c) shows calculated transition rates between states of different manifolds. For initial states in manifolds  $n \geq 2$ , decay can occur over a wide spectral range, while decay from  $n = 1$  states leads to a series of discrete emission lines, with the first excited transition  $\sim 30$  meV (21 nm) blue detuned from the  $X^0$  line (not visible in Fig. 1(c)). We note that similar excitation spectra exist for a charged QD. In contrast to the  $X^0$  level spectrum, here also the  $n = 0^\pm$  manifolds consist of multiple states owing to the excited confined motional states of the extra charge. Hence, for charged QDs cavity feeding can also occur for  $1^\pm \rightarrow 0^\pm$  transitions.

Due to the excited-state nature of cavity feeding for a neutral QD, we expect a super-linear pump-power dependence of the CM emission. In Fig. 2(a) we show the integrated intensities of the CM together with those of the  $X^0$ ,  $X^{1-}$ , and  $XX^0$  lines as a function of pump power in a log-log plot. As expected, the  $X^0$  and  $X^{1-}$  lines follow an approximately linear power dependence below saturation (which is defined through the emission maxi-

imum of the respective line). A power-law fit to the  $XX^0$  data gives an exponent of  $\sim 1.5$ . In contrast, the dynamics of the CM is more complex. For pump powers below  $\sim 100$  nW, the CM follows a linear dependence, similar to the  $X^0$  and the  $X^{1-}$ . This observation is consistent with our model since for very low pump powers we expect the charged  $1^\pm \rightarrow 0^\pm$  exciton transitions to give the dominant contribution to cavity feeding. When increasing the pump power, the behavior of the CM emission becomes super-linear: in between the two vertical lines in Fig. 2(a), the cavity displays the power dependence of the  $XX^0$  line. Furthermore, the cavity intensity increases far above the saturation levels of both the  $X^0$  and the  $XX^0$ . For powers above 500 nW, the cavity luminescence follows that of the QD p-shell as clearly demonstrated in Fig. 2(b). Here we recorded PL data over a wide spectral window covering the QD s-, p- and d-shells and plot the total integrated intensity of the shell emissions as a function of pump power. Since p- and d-shell emission originate from higher-excitation manifolds of the QD [17], these observations strongly support our model of cavity feeding.

As argued earlier, the most striking feature of cavity feeding is the peculiar interplay of quantum correlations observed in off-resonant cavity PL [3, 4]. To obtain a better understanding of these observations, we carried out pump-power dependent photon correlation measurements that show new features that were not visible in earlier measurements. Figure 3(a)–(c) display normalized CM- $X^0$  cross-correlation curves for a mutual detuning of about 10.3 meV (7.3 nm) for three different pump powers. The histograms were taken at pump powers of about (c) 20%, (b) 40% and (a) 80% of the saturation power  $P_{\text{sat}} \approx 600$  nW. For all three pump powers, there is a strong suppression of coincidences for zero time delay. In other words, CM and  $X^0$  emission do not occur simultaneously, which follows naturally from our model since cavity feeding arises from QD transitions other than the  $X^0$ . Moreover, we observe an abrupt turn-on of  $X^0$  luminescence upon detection of a cavity photon, reflected by the bunching feature for  $\tau > 0$ . This can easily be explained by the fact that for pump powers below saturation a significant portion of cavity photons is emitted in  $2 \rightarrow 1$  transitions. In this case, detection of a cavity photon projects the QD into the  $n = 1$  manifold, leading to a higher-than-average probability for  $X^0$  emission. When increasing the pump power, the average QD population is higher, such that the detection of a cavity photon does not increase the conditional probability that the QD is in the  $n = 1$  manifold. This is manifested in a reduction and eventually the disappearance of the bunching peak for higher powers, as clearly visible in Figs. 3(a) and (b).

Surprisingly, in our previous study the cavity photons did not show any significant quantum correlations [3] even though the cavity is fed by a single quantum emitter. To gain further insight, we measure the second-

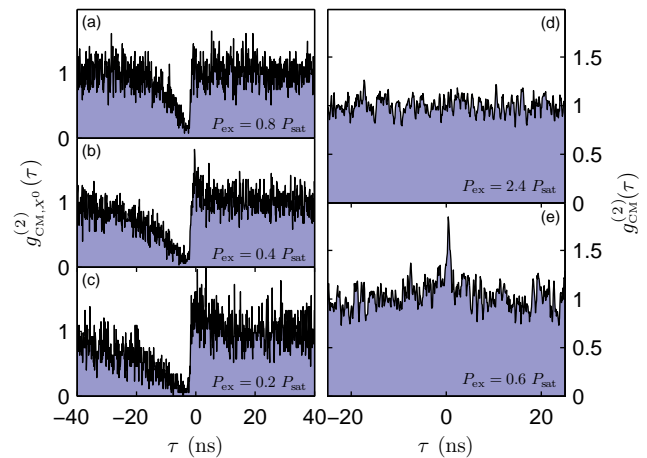


FIG. 3: (color online) Experimental correlation histograms. (a)–(c) Cross-correlations between the CM and the  $X^0$  for different pump powers.  $\tau > 0$  refers to  $X^0$  emission upon detection of a cavity photon. The bunching feature for  $\tau > 0$  disappears when increasing the pump power, as expected from our model. The timescale for the anti-bunching feature for negative delays reflects the re-pump time of the system. (d)–(e) CM auto-correlations for two different pump powers. Below saturation, clear bunching at  $\tau = 0$  is visible which disappears above saturation, in agreement with the model.

order auto-correlation function  $g_{\text{CM}}^{(2)}(\tau)$  of the cavity luminescence for different pump powers. Here, the cavity is detuned by 15.3 meV (10.9 nm) to the red of the  $X^0$ . Figure 3(d) and (e) display the outcome for two different regimes of pump powers. While for a pump power  $P_{\text{ex}} \approx 0.6 P_{\text{sat}}$  [Fig. 3(e)] the cavity emission exhibits clear bunching for  $\tau = 0$ , well above saturation the bunching disappears and the correlations are purely Poissonian [Fig. 3(d)]. We argue that the observation of bunching, not observed in earlier single QD experiments, provides a strong indication of cavity feeding from QD excited states as predicted by our model: when the system is driven well below saturation, the likelihood of finding the QD in a  $n \geq 2$  manifold is very small. Detection of a cavity photon however, increases the conditional probability of finding the QD in a higher manifold, implying in turn that a second photon-emission event is more likely than on average. In contrast, for higher pump powers the average population of excited states is higher and a detection of a cavity photon does not increase the likelihood of a second photon detection event, since the emission of a cavity photon can arise from transitions between any two neighboring excitation manifolds ( $n \rightarrow n - 1$  with  $n = 2, 3, \dots$ ). In this regime, cascaded cavity-photon emission leads to Poissonian statistics.

In order to demonstrate the consistency of our experimental findings and our model, we determine the transition rates between the calculated eigenstates by considering coupling to LO phonons, cavity coupling and spin-flip processes. The dynamics of the system is then studied by

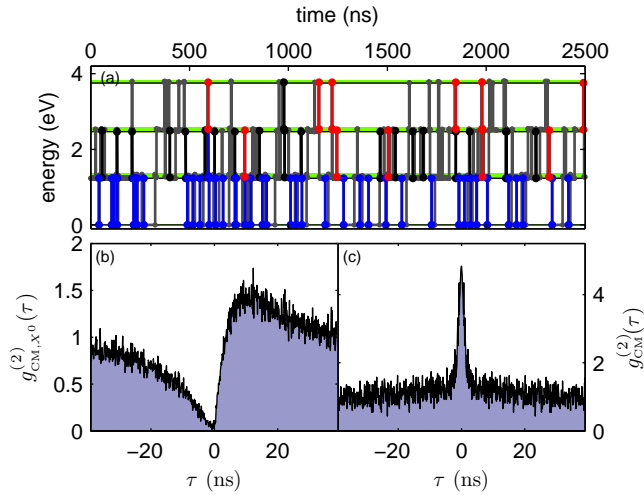


FIG. 4: (color online) Simulation of cavity-feeding dynamics. (a) Monte-Carlo random-walk of excitation and photon-emission events (gray lines). Transitions resonant with the CM,  $X^0$ , and  $XX^0$  lines are depicted in red, blue, and black, respectively. (b) Extracted CM- $X^0$  cross-correlation histogram reproducing the characteristic features observed in the experiment (compare Fig. 3(c)). (c) Extracted CM auto-correlation curve, exhibiting strong bunching at zero time delay in accordance with the experimental result of Fig. 3(e).

performing a Monte-Carlo random walk for a given excitation rate [18, 19]. A typical result depicting the quantum jumps occurring in the system is shown in Fig. 4(a). The events taking place at the cavity-mode frequency are indicated by red lines, whereas  $X^0$  and  $XX^0$  events are shown in blue and black, respectively. Clearly, all cavity photons here are emitted in  $2 \rightarrow 1$  and  $3 \rightarrow 2$  transitions; in contrast, there are no cavity photons emitted in  $1 \rightarrow 0$  transitions, since there cavity feeding is energetically not allowed, in perfect agreement with the arguments given above. We remark that our simulation does not include cavity feeding from charged excitonic compounds. However, including charged exciton states would not affect the general validity of the model or our findings.

From the simulated Monte-Carlo random walk, we extract both cross-correlation and auto-correlation histograms which are displayed in Fig. 4(b) and (c). In the simulation a CM detuning of 7 meV (5 nm) red of the  $X^0$  was assumed [24]. In general, the experimental observations of Fig. 3 are well reproduced by the simulations. The cross-correlation in Fig. 4(b) exhibits the two main features already observed in the experiment: anti-bunching at zero time delay and a bunching peak for positive times [25]. Figure 4(c) shows a cavity auto-correlation trace for a pump power below saturation. As expected from our qualitative arguments, the curve shows strong bunching with  $g_{CM}^{(2)}(0) \approx 4.8$ . In the experiment, however, we only measure  $g_{CM}^{(2)}(0) \approx 1.8$ , which we attribute to two effects: first, the experimental pump

power is difficult to relate to the theoretical excitation rate and  $g_{CM}^{(2)}(\tau)$  strongly depends on the pump power. Second, in the experiment cavity feeding can additionally arise from charged  $1^\pm \rightarrow 0^\pm$  transitions. Consequently, we expect a reduction of bunching, since here an  $n \geq 2^\pm$  manifold excitation is not required for cavity feeding.

In conclusion, we have developed a model for cavity feeding based on cascaded QD emission from excited states of multi-exciton manifolds. In particular, the peculiar photon-correlation features observed experimentally can fully be accounted for in our model. Moreover, the model is valid for a large range of detunings — particularly in the far-detuned case for which acoustic phonon mediated feeding cannot play a role. The work presented here allows for a more refined understanding of QD-based implementations of cavity QED and the limitations of future spin-photon interfaces such devices are predicted to realize. Moreover, it could have strong implications for experiments on lasing [20, 21] in such systems. An interesting extension of the present work could be the investigation of feeding in charge-controlled devices [22].

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  - [23] We remark here that stability of triplet states in the  $n \geq 2$  manifolds against LO-phonon relaxation is likely to play a key role in enhancing cavity feeding from the excited orbital states of the corresponding manifold.
  - [24] The truncated continuum of wetting-layer states leads to

non-physical results when going to larger detunings.

- [25] Here the bunching feature for  $\tau > 0$  is delayed compared to the experimental trace, which could partly be due to

the simplified model used for the spin-flip process.